

## P ENT COOPERATION TREA

PCT

NOTIFICATION CONCERNING  
AMENDMENTS OF THE CLAIMS(PCT Rule 62 and  
Administrative Instructions, Section 417)

From the INTERNATIONAL BUREAU

To:

Assistant Commissioner for Patents  
United States Patent and Trademark  
Office  
Box PCT  
Washington, D.C. 20231  
ETATS-UNIS D'AMERIQUE

in its capacity as International Preliminary Examining Authority

Date of mailing (day/month/year)

07 August 2000 (07.08.00)

International application No.

PCT/US99/28038

International filing date (day/month/year)

24 November 1999 (24.11.99)

Applicant

BOARD OF REGENTS OF UNIVERSITY OF NEBRASKA et al

The International Bureau hereby informs the International Preliminary Examining Authority that no amendments under Article 19 have been received by the International Bureau (Administrative Instructions, Section 417).

The International Bureau of WIPO  
34, chemin des Colombettes  
1211 Geneva 20, Switzerland

Facsimile No. (41-22) 740.14.35

Authorized officer

Olivia RANAIVOJAONA

Telephone No. (41-22) 338.83.38

## P. ENT COOPERATION TREA

PCT

## NOTIFICATION OF ELECTION

(PCT Rule 61.2)

From the INTERNATIONAL BUREAU

To:

Assistant Commissioner for Patents  
United States Patent and Trademark  
Office  
Box PCT  
Washington, D.C.20231  
ETATS-UNIS D'AMERIQUE

in its capacity as elected Office

Date of mailing (day/month/year) 07 August 2000 (07.08.00)	
International application No. PCT/US99/28038	Applicant's or agent's file reference UNVN62457/05
International filing date (day/month/year) 24 November 1999 (24.11.99)	Priority date (day/month/year) 25 November 1998 (25.11.98)
Applicant DOWBEN, Peter, A. et al	

1. The designated Office is hereby notified of its election made:

☒ in the demand filed with the International Preliminary Examining Authority on:  
21 June 2000 (21.06.00)

☐ in a notice effecting later election filed with the International Bureau on:  
\_\_\_\_\_

2. The election ☒ was  
☐ was not

made before the expiration of 19 months from the priority date or, where Rule 32 applies, within the time limit under Rule 32.2(b).

The International Bureau of WIPO 34, chemin des Colombettes 1211 Geneva 20, Switzerland Facsimile No.: (41-22) 740.14.35	Authorized officer Olivia RANAIVOJAONA Telephone No.: (41-22) 338.83.38
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**BORON-CARBIDE SOLID STATE NEUTRON DETECTOR AND**  
**METHOD OF USING SAME**

**STATEMENT REGARDING FEDERALLY SPONSORED RESEARCH OR  
DEVELOPMENT**

- 5           The Board of Regents of the University of Nebraska acknowledges that some funding for the research leading to this application was provided by the United States Government.

**CROSS REFERENCE TO RELATED APPLICATIONS**  
Not Applicable.

10                           **BACKGROUND OF THE INVENTION**

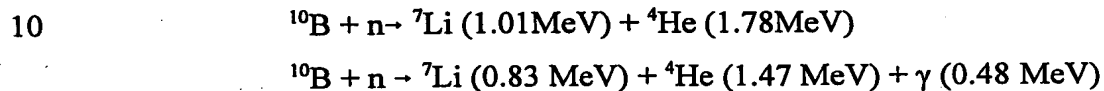
The present invention relates to detection of neutrons. More specifically, the present invention relates to a method and device for the efficient detection of neutrons that employs a boron-rich semiconductor as an electrically active part of the detection device.

- 15           Neutron scattering is an important research method to determine the structure of solids and liquids. It is used to understand the forces that act between the atoms in these systems and to determine the magnetic behavior of materials as well. The research and practical applications cover a broad range of areas, from the basic properties of materials to studies of engineering and medical applications.

- 20           There are essentially only four elements suitable for forming solid state semiconductor neutron detectors – boron (B), cadmium (Cd), gadolinium (Gd) and lithium (Li). Lithium semiconductor materials exist ( $\text{LiInS}_2$ ,  $\text{LiInSe}_2$  and  $\text{LiZnP}$ ) but are difficult to reliably fabricate into devices and are very difficult materials with which to work. Gadolinium conversion layer based silicon (Si) diodes have been fabricated and  
25   proposed for neutron detection, but are not particularly stable. Cadmium zinc telluride has been shown to yield thermal neutron detection and the cadmium neutron capture cross section is high, but the neutron capture produces such high energy gamma rays (over 0.5

MeV) that the detectors would have to be large in order to detect these gammas efficiently.

Use of boron with neutron detectors is known both in the scintillator, the gas and the conversion layer varieties. Boron phosphide (BP) heterojunction diodes with silicon were successfully tested as alpha radiation detectors, but failed to work as neutron detectors. Boron carbide ( $B_4C$ ) was successfully used as a neutron detector based upon resistivity changes resulting from increased lithium doping, as were (111) BP wafers. The lithium production in the boron carbide was a result of the following nuclear reactions:



Boron has also been considered as a coating to a silicon diode and a GaAs diode but the maximum efficiency is low (less than 5%).

Existing gas and liquid neutron detectors are much larger and less rugged than solid-state ones could be. However, existing solid state neutron detectors also suffer serious limitations. For example, known boron-doped semiconductors are only a few percent efficient because they contain relatively little boron. Gadolinium, lithium and hydrocarbon conversion layers are all adversely affected by corrosion and high temperatures.

20                   Furthermore, known conversion layer devices have low efficiencies, unless multiply stacked, because the range of the reaction products in the material of the conversion layer is generally considerably less than the thickness required for stopping thermal neutrons. Gadolinium conversion layers are an exception - but the neutron - gadolinium reaction results in conversion electrons of relatively low energy (70 keV) compared with the reaction products in the case of neutron capture by boron 10. Cadmium zinc telluride has been shown to yield thermal neutron detection, but the neutron capture produces such high energy gamma rays (over 0.5 MeV) that the detectors must be large to detect these gammas efficiently. Scintillator combinations with photomultipliers or intensified cameras are bulky and heavy and, except for neutron-

detecting scintillating fibers coupled optically to a remote photomultiplier or camera, are intolerant of high temperatures.

Boron and boron compounds, including boron carbide, are also used in neutron absorbing shielding purposes in nuclear reactors and other types of neutron radiation environments. For example, boron carbide can be used with shielding, thermal electric power, or detection of neutrons (by means of the resistivity change not by detection of individual neutrons). However, use of boron carbide to detect neutrons where the boron carbide is an electrically active semiconductor is novel.

### SUMMARY OF THE INVENTION

10 It is an object of the present invention to provide an inexpensive solid state neutron detector that includes a robust, structurally forgiving boron rich semiconductor.

It is another object of the present invention to provide a boron carbide semiconductor that utilizes its electrical properties as a semiconductor rather than its electrical property of resistance as a means of detecting neutrons or its thermoelectric properties in detecting neutrons.

15 A still further object of the present invention is to provide a detection device that yields high gain.

A further object of the present invention is to provide a detection device that provides real time response.

20 A further object of the present invention is to provide a detection device that is capable of detecting single neutrons.

Yet another object of the present invention is to provide a detection device that has low sensitivity to gamma and other radiation.

25 Still another object of the present invention is to provide a method of detecting neutrons with a detector device having a boron carbide semiconductor.

According to the present invention, the foregoing and other objects are obtained by a detection device having a layer of boron carbide. In the device, the boron carbide layer is an electrically active part of the detection device. The sensing mechanism of the detection device is inherent in the electrically connected, semiconducting boron carbide layer, which provides neutron capture resulting in prompt,

innately highly amplified, electrical output signals following interception of neutron(s).

Additional objects, advantages, and novel features of the invention will be set forth in part in the description which follows, and in part will become apparent to those skilled in the practice of the invention. The objects and advantages of the invention  
5 may be realized and attained by means of the forms of instrument and the combinations particularly pointed out in the appended claims.

#### BRIEF DESCRIPTION OF THE SEVERAL VIEWS OF THE DRAWINGS

In the accompanying drawings which form a part of the specification and which are to be read in conjunction therewith and in which like reference numerals are  
10 used to indicate like parts in the various views:

Fig. 1 is a schematic representation of a heterojunction diode embodying the present invention;

Fig. 2 is a schematic representation of the test device using the principles of the present invention.

15 Fig. 3 depicts voltage-current characteristics of heterojunction diodes of the present invention;

Fig. 4 depicts count rates of neutrons with insertion of heterojunction diodes of the present invention into a neutron reactor; and

20 Figs. 5 and 6 depict the relationship of ideally attainable neutron detection efficiency as a function of the thickness of the boron-carbide layer of heterojunction diodes of the present invention in the cases of natural 10 Boron abundance and 100% 10 Boron enrichment of the boron carbide layer.

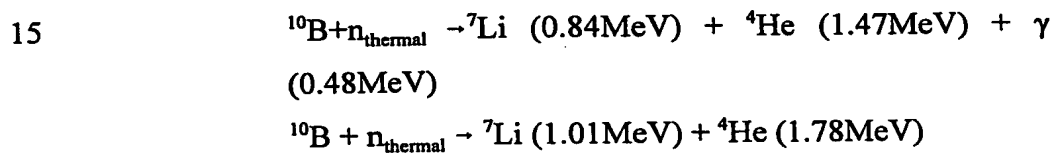
#### DETAILED DESCRIPTION OF THE INVENTION

Referring initially to Fig. 1, a heterojunction diode 10 is shown. This  
25 invention also applies to homojunction diodes and other known semiconductor detection devices, examples of which are provided below. Diode 10 is shown as having a boron carbide boron-carbon alloy semiconductor 12 on a silicon substrate 14. Semiconductor 12 is grown by plasma-enhanced chemical vapor deposition (PECVD). The preferred deposition technique is disclosed in U.S. Patent Nos. 4,957,773 (Spenser, et al.);  
30 5,468,978 (Dowben); 5,658,834 (Dowben), which patents are expressly incorporated by reference herein. A pair of sputter-deposited gold electrodes 16 communicate with

semiconductor 12 and substrate 14. Secured to each electrode 16 is a wire 18 that serves to connect electrodes 16 to a bias voltage source and an electrical detection device such as a charge pulse measurement circuit. The sensory/measurement devices as well as monitoring devices are known and will not be discussed further.

Essentially, the invention works by including a boron-rich carbon alloy as an electrically active semiconductor region of a detector and by placing the detector where it can receive neutrons. The preferred way to detect neutrons is with atoms which are the most likely to capture neutrons and in which each neutron capture leads to the creation of one or more energetic charged particles whose mass is large compared with that of an electron and whose energy is large and can efficiently be converted to a measurable electrical signal. Boron atoms are highly likely to capture neutrons and such neutron capture creates highly energetic ions.

The following two reactions between boron isotope 10 ( $^{10}\text{B}$ ) and a thermal neutron form the basis for neutron detection as contemplated by the present invention:



20 With a boron-rich semiconductor, the boron captures the neutron and promptly decays into high-energy ions. The energetic ions cause secondary ionization of orders of magnitude more atoms in the surrounding materials for each captured neutron, liberating a correspondingly large electrical charge. The diode nature of the device enables the electrical charge to be collected. Also, incorporating the boron-rich alloy as an electrically active semiconductor part of the detector allows for the overall thickness of the device to be reduced while retaining high efficiency of neutron detection.

25           The first device to use this concept was a boron-carbon alloy semiconductor (grown by plasma-enhanced chemical vapor deposition) on a silicon substrate with sputter-deposited gold electrodes, as shown in Fig. 2. As seen in Fig. 2, a boron carbide/silicon diode 20 is connected to a charge sensitive preamplifier 22.

Charge sensitive preamplifier 22, in turn, is connected to a bias voltage input 24 and a single channel analyzer / multichannel scaler 26 which is connected to a computer 28.

In this heterojunction diode, the above reactions lead to dense local ionization of atoms and hence production of electron-hole pairs (at least of order  $5 \times 10^5$  pairs per neutron reaction), many of which are collected due to the applied bias voltage and form a charge pulse which is registered and counted by external circuitry. Such a device was first tested successfully on July 24, 1998 at the nuclear reactor in the VA Hospital in Omaha, Nebraska. This device could be improved in several ways, including  $^{10}\text{B}$ -enrichment (to nearly 100%  $^{10}\text{B}$  from the naturally occurring approximately 19%  $^{10}\text{B}$  found in unenriched boron), increasing the thickness and quality of the boron carbide layer, changes in the electrical configuration and electrical circuitry, and changes in the functional and geometrical configurations.

The deposition of films for the heterojunction diodes (boron-carbon alloy,  $\text{B}_3\text{C}$ , on (111) Si) performed in this test was undertaken in a custom designed parallel plate 13.56 (MHz radio-frequency PECVD reactor used in previous studies). The silicon substrates were doped to  $7 \times 10^{14}/\text{cm}^3$ . The (111) Si substrates surfaces were prepared by  $\text{Ar}^+$  ion sputtering in the plasma reactor. The source molecule gas closo-1,2-dicarbododecaborane (ortho-carborane,  $\text{C}_2\text{B}_{10}\text{H}_{12}$ ) was used as the source compound for growing the boron carbon alloy.

Typical  $\text{B}_3\text{C}/\text{n-type}$  silicon heterojunctions have been routinely formed by this technique. An example of one such diode device is presented in Fig. 2 with the boron carbide alloy layer of about 1000 nm thick as used as a neutron detector. These devices typically have onsets of 1 eV with very little leakage current (less than 5  $\mu\text{A}$  at  $25^\circ\text{C}$ ) and the boron carbide layer has the p-type character of the undoped PECVD semiconducting boron carbide in this device topology.

The detector area of these heterojunction diodes was about  $1 \text{ cm}^2$ , and wired in a "mesa" geometry. The neutron source was a small TRIGA-type reactor (V.A. Medical Center, Omaha, NE) with a flux of  $1.6 \times 10^6 \text{ n/cm}^2 \cdot \text{s}$  based on calculations for the fission chamber. A heterojunction diode, reversed biased to about 3 V, was wired for pulse counting as shown in Figure 2 and inserted into the reactor. The resulting count



rates with insertion are plotted in Figure 3. Background and noise counts are in the range of 250 to 300 Hz, and within the reactor, the count rate rises to  $2 \times 10^5$  Hz.

To assure that very little of this count rate is attributable to gamma radiation, the diode was tested against a 100 mCi  $^{137}\text{Cs}$  source for gamma radiation at a distance of 10 cm. The 661 keV gamma rays provided no detectable increase in count rate above background in spite of an expected  $10^6$  gamma rays incident on the diode per second. This is consistent with the expected extremely low gamma-ray sensitivity of such a solid state boron-carbon/silicon semiconductor alloy device, since boron and carbon have low atomic numbers and the boron-rich detectors were made very thin (1000 nm), and the electrically active silicon layer was under 600 nm thick.

Given that almost all counts are attributable to neutrons and that the boron carbide film is about 1000 nm thick, the detection efficiency is thus about 1%. Given that devices can be made with boron carbide of 50 nm to 100 nm in thickness and with depletion layers extending several nanometers, the single (thermal) neutron detection efficiencies are, conservatively, expected to reach 80% and higher in devices which simultaneously have exceedingly low  $\gamma$ -ray sensitivity ( $< 1\%$  detection efficiency for all energies greater than 100 keV and  $< 0.01\%$  for all energies above 0.5 MeV, assured by the use of boron as the dominant atomic species). Since the neutron -  $^{10}\text{B}$  interaction results almost exclusively in the yield of highly ionizing lithium ions and alpha particles of total kinetic energy about 1.5 MeV and the boron atoms form the major species in the active semiconducting regions of the devices, the boron-carbon alloy layer of the detector yields an enormous internal gain (considerably greater than  $10^5$ ) which is essentially noise-free and comparable with the gain of the intensifiers and photomultipliers commonly used in scintillation-based detectors and imagers. By using exclusively  $^{10}\text{B}$  enriched boranes in the PECVD fabrication process, detection efficiency with thinner films can be considerably improved compared with devices whose  $^{10}\text{B}$  content reflects the natural isotopic abundance, about 19%  $^{10}\text{B}$ .

As seen in Figure 2, the electronics demands are minimal compared with those for gadolinium neutron conversion layer-based detectors (which rely on the much smaller 70 keV energetically available for signal generation by the conversion electrons from gadolinium), while ensuring considerably greater efficiency and stability.

Additionally, the boron-carbon devices can be thinner than 100  $\mu\text{m}$  thick and still achieve nearly 100% thermal neutron detective efficiency. Stacking diodes, interleaved with neutron energy absorbers, to form efficient neutron "calorimeters" or spectrometers is also possible. In combination with boron carbide based high temperature electronics, the boron-carbon based neutron detection systems are expected to be particularly applicable in harsh environments because of the refractory and mechanical performance of boron carbide. The boron-carbon devices may even be fabricated on metal substrates as well as fabricated with spatial resolution that could be on scales smaller than 0.5 nm. There is the possibility of fabricating spatial array detectors, including position sensors for scattering experiments, as well.

High efficiency is achieved because there is a proportionally large amount of boron present in the semiconductor layer. The boron carbide semiconductor has boron of whatever isotope one therefore chooses present in atomic fractions in the order of 80%. This is exceedingly rich in boron compared with any other suitable semiconductor. Because the density of boron atoms in the material is so high, the boron-rich layer can be quite thin and still contain enough boron atoms per unit area to be able to detect the neutrons very efficiently. In naturally occurring boron there is close to 20% of the boron atoms which are  $^{10}\text{B}$  atoms which are the isotopes which interact strongly with neutrons to give the reactions provided above. It is certainly possible to increase the fraction of boron that is  $^{10}\text{B}$  from natural abundance to about 95% or higher. This enrichment would result in  $^{10}\text{B}$  atoms accounting for a fraction, about 80% or higher, of all atoms in the semiconductor boron carbide layer. Hence, if material enriched in  $^{10}\text{B}$  is used rather than just the naturally occurring isotope ratio of  $^{10}\text{B}$ , the efficiency increases even further.

Another important issue for efficiency is not just the reaction of the neutron with boron, but the ability to detect the reaction. By incorporating the boron atoms in an electrically active semiconductor where the lithium ion and the alpha particle can cause dense ionization of other atoms, many electron-hole pairs can be created by ionization of the atoms, and the electric fields that can be applied across the boron carbide layer can sweep out a large fraction of the electron-hole pairs. Thus, there are three aspects to efficiency. The first is  $^{10}\text{B}$  being present in large number density. The second

being that the reaction of  $^{10}\text{B}$  with neutrons results in ions which very efficiently ionize atoms in the surrounding in an electrically active semiconductor where the charge can be swept out efficiently. The third aspect of efficiency is that  $^{10}\text{B}$  results in ions which have such a large energy that they can produce very large numbers of detectable electron hole  
5 pairs. The reactions which occur between neutrons and the other elements which give probable neutron interactions don't result in reaction products which are as readily detectable or detectable to give such large signals. Boron is unique.

Another point concerns detection devices having conversion layers containing boron. Neutron capture by boron generates the alpha particle and the lithium  
10 ion which can only travel a very limited distance. If conversion layer contained enough boron atoms to cause capture of a sufficient fraction of neutrons, then the layer will be so thick that the lithium and the alpha particles in some cases will not get out of the boron layer and, therefore, will not generate signals that are readily detectable. This is a severe defect compared with the boron carbide semiconductor devices of the present invention.

15 This invention can be used in various forms of solid-state neutron detectors presenting entrance detecting areas of order  $\mu\text{m}^2$  to  $\text{m}^2$ . These detectors are capable of being implemented with very thin detecting and electrically active regions ( $\leq 1 \mu\text{m}$  minimum effective electrical thickness), with very low mass per unit detecting area, with efficiencies ranging up to nearly 100% even for single neutrons, with real-time  
20 response, with high spatial resolution ( $\leq 1 \mu\text{m}$  minimum), and with high temporal resolution. Of course, implementation may not always need to, or be able to, employ each of these attributes. Voltage and power needs are slight, as are charge pulse processing requirements.

Although the invention is described above as relating to heterojunction  
25 diodes, it is to be understood that the invention can be implemented in a large number of other ways, including homojunction diodes; p-i-n diodes; metal-semiconductor-metal, Schottky and other diodes; transistors; diode and transistor arrays; charge-induced devices (CID) and CID arrays; charge-coupled devices (CCD) and CCD arrays; solid-state neutron-detecting analogs of "photomultipliers"; neutron semiconductor avalanche  
30 devices; position-sensitive detectors, including those relying on charge subdivision or sensing and on current subdivision and those having capacitive or resistive means of

doing so; semiconductor drift detectors or semiconductor drift chambers; stacked series of one or more of the above detector types which are configured to serve as neutron energy spectrometers; individual or stacked series of one or more of the above detector types which also, or alternatively, serve as dosimeters. The dosimeters can be capable of yielding both real-time and cumulative dosimetry information once or many times, completely nondestructively of the dosimetry information contained in the detectors.

The range of applicability of the present invention includes: medical radiation dosimetry; detecting nuclear material; anti-terrorism and anti-smuggling devices; monitoring of nuclear reactors, of nuclear storage units and facilities, and of nuclear weapons, weapons storage and weapons shipment; life science materials and physical sciences scattering experiments; monitoring of neutron sources; calibration of neutron flux; personnel and environmental radiation protection; radiation protection at high energy radiation facilities, including medical x-ray facilities (high energy ones); neutron cancer therapy; profiling of medical, therapeutic, research and other neutron beams; comet, planetary and other space exploration.

From the foregoing, it will be seen that this invention is one well adapted to attain all the ends and objects herein above set forth together with other advantages which are obvious and which are inherent to the structure. It will be understood that certain features and subcombinations are of utility and may be employed without reference to other features and subcombinations. This is contemplated by and is within the scope of the claims.

Since many possible embodiments may be made of the invention without departing from the scope thereof, it is to be understood that all matter herein set forth or shown in the accompanying drawings is to be interpreted as illustrative and not in a limiting sense.

Having thus described the invention, what is claimed is:

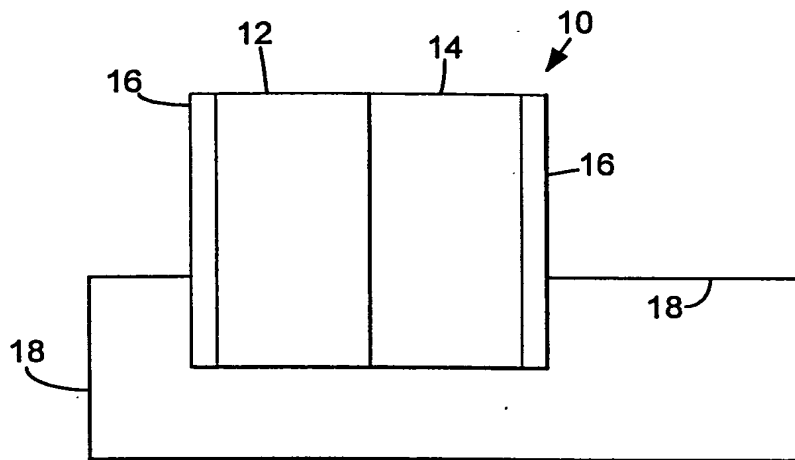
1. A neutron detection device, said device comprising: a sensing mechanism, said sensing mechanism having a layer of boron carbide semiconductor wherein the boron carbide layer is an electrically active part of said detection device; and a monitoring device, wherein said monitoring device records said changes in said boron carbide layer detected by said sensing mechanism.
2. The device of claim 1, wherein said sensing mechanism is inherent in said boron carbide semiconductor layer and results in a prompt, innately highly amplified, electrical output following capture of a single neutron.
3. The device of claim 2, wherein said device is a homojunction diode.
4. The device of claim 1, further comprising a layer of silicon communicating with said layer of boron carbide.
5. The device of claim 4, wherein said device is a heterojunction diode.
6. The device of claim 1, wherein the thickness of said boron carbide layer is about 1000 nm.
7. The device of claim 5, wherein the thickness of said silicon layer is less than 600 nm.
8. The device of claim 1, further comprising at least two diodes interleaved with a neutron energy absorber.
9. The device of claim 1, wherein said boron carbide layer is fabricated on a metal substrate.
10. The device of claim 1, wherein said boron carbide layer contains at least 80%  $^{10}\text{B}$ .
11. The device of claim 1, wherein said device is capable of operating at 500 °C.
12. A method of detecting neutrons, said method comprising: positioning a neutron detecting device in a location to allow said device to intercept a stream of neutrons, said detector comprising a layer of boron carbide wherein said boron carbide layer is an electrically active part of said device, and a sensing mechanism coupled to said boron carbide layer; introducing at least one neutron traveling in a direction to be intercepted by the boron carbide layer; and monitoring the interaction of the neutron with

the boron carbide semiconductor; wherein said sensing mechanism detects changes in said boron carbide layer caused by the interception of neutrons.

13. A method of detecting neutrons, said method comprising: positioning a neutron detecting device in a location to allow said device to intercept a stream of neutrons, said detector comprising a layer of boron carbide wherein said boron carbide layer is an electrically active part of said device, and a sensing mechanism inherent to said boron carbide layer; introducing at least one neutron traveling in a direction to be intercepted by the boron carbide layer; and monitoring the interaction of the neutron with the boron carbide semiconductor; wherein said sensing mechanism detects changes in said boron carbide layer caused by the interception of neutrons.

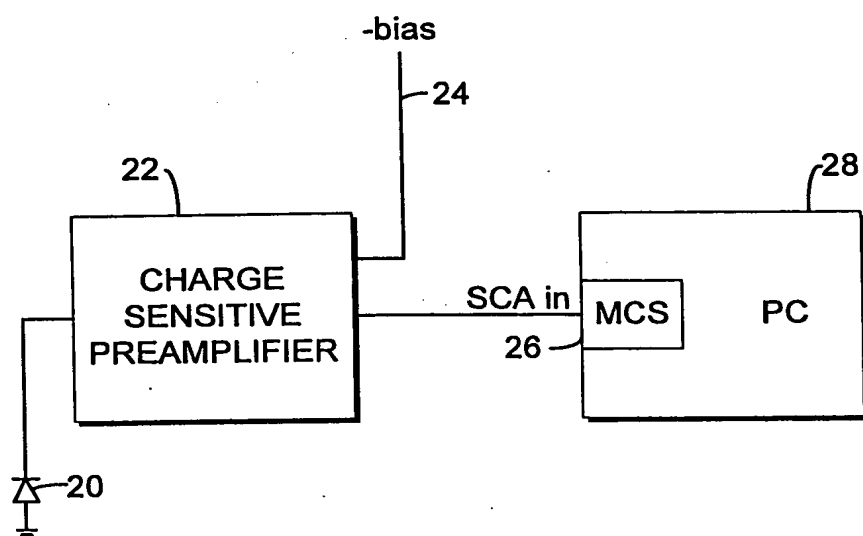
### **ABSTRACT OF THE DISCLOSURE**

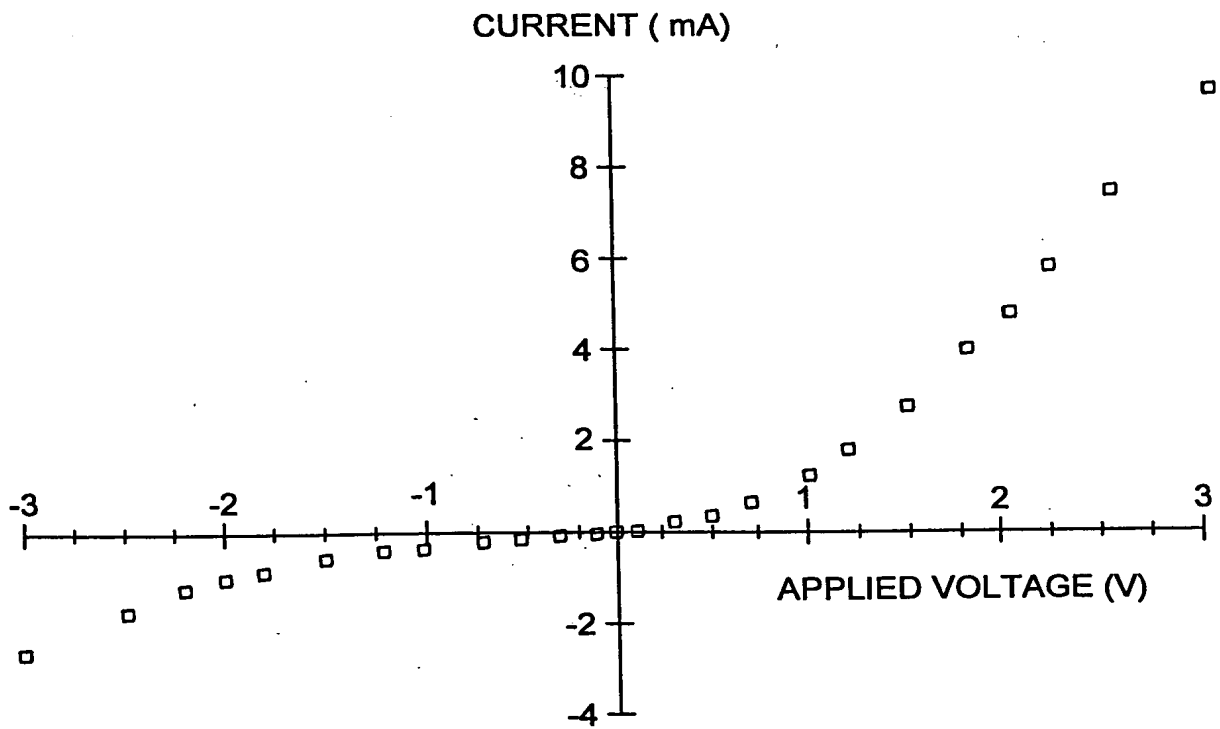
A boron carbide solid state neutron detector and method of using the detector is disclosed, wherein the detector includes a layer of boron carbide wherein the boron carbide layer is an electrically active part of the detection device, a sensing mechanism inherent to said boron carbide layer, wherein the sensing mechanism detects changes in the boron carbide layer caused by the interception of neutrons and a monitoring device coupled to the sensing mechanism.

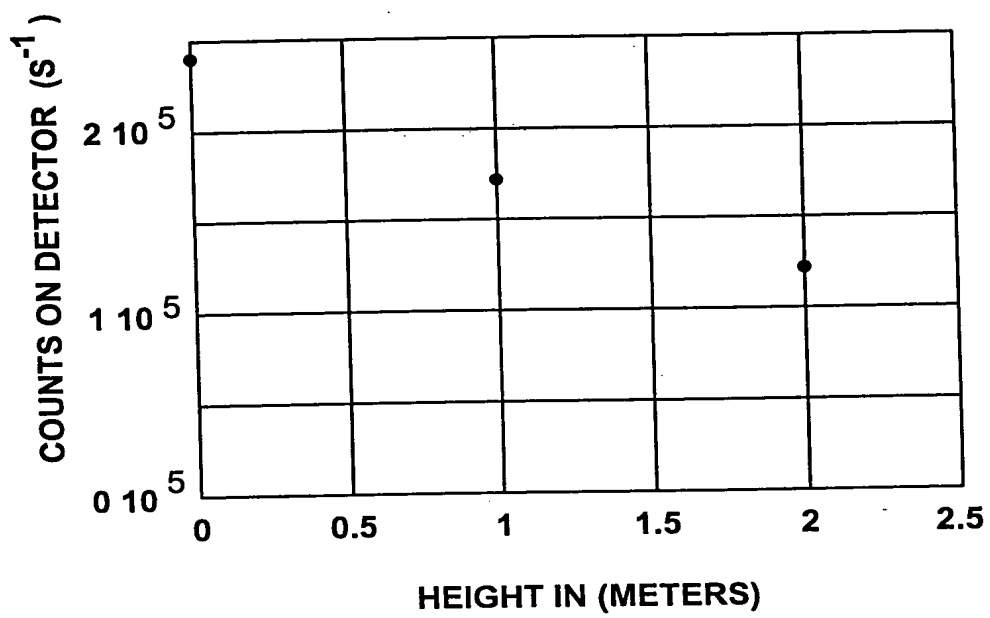


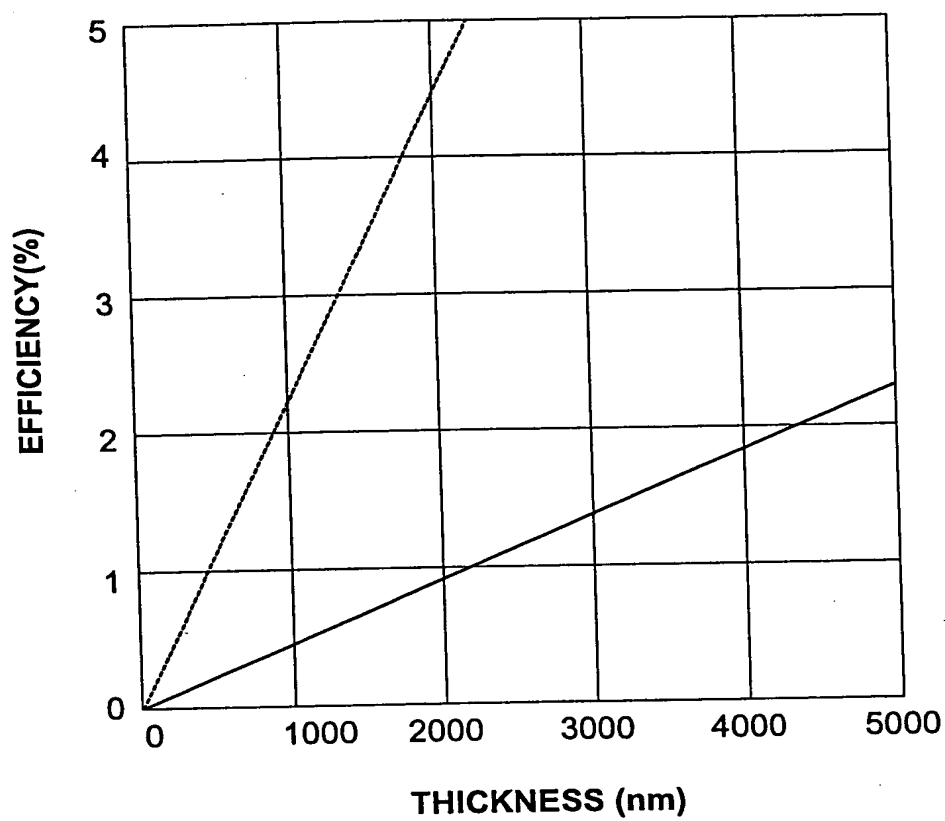
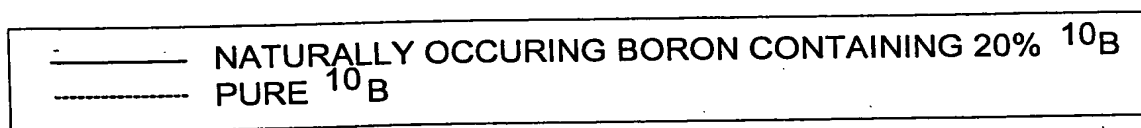
**FIG.1.**

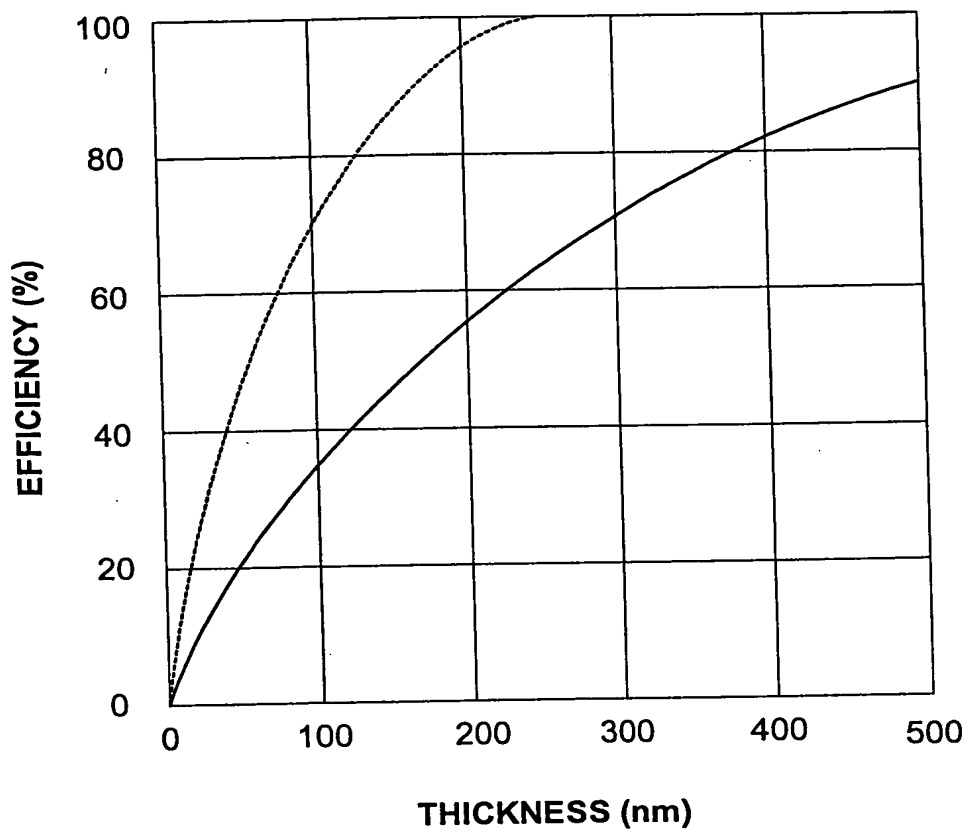
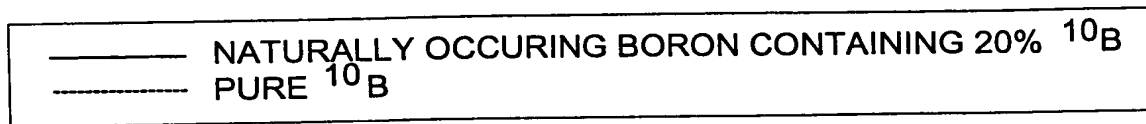


**FIG.2.**

**FIG.3.**

**FIG. 4.**

**FIG. 5.**

**FIG. 6.**

## PATENT COOPERATION TREATY

## PCT

## INTERNATIONAL PRELIMINARY EXAMINATION REPORT

(PCT Article 36 and Rule 70)

REC'D 28 DEC 2000

WIPO

PCT

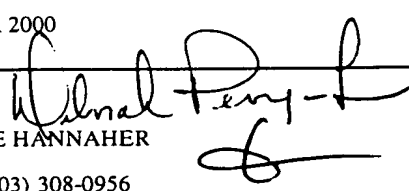
Applicant's or agent's file reference UNVN62457/05	<b>FOR FURTHER ACTION</b> See Notification of Transmittal of International Preliminary Examination Report (Form PCT/IPEA/416)	
International application No. PCT/US99/28038	International filing date (day/month/year) 24 NOVEMBER 1999	Priority date (day/month/year) 25 NOVEMBER 1998
International Patent Classification (IPC) or national classification and IPC IPC(7): G01T 3/08 and US Cl.: 250/370.05		
Applicant BOARD OF REGENTS OF UNIVERSITY OF NEBRASKA		

1. This international preliminary examination report has been prepared by this International Preliminary Examining Authority and is transmitted to the applicant according to Article 36.
2. This REPORT consists of a total of 4 sheets.  
☒ This report is also accompanied by ANNEXES, i.e., sheets of the description, claims and/or drawings which have been amended and are the basis for this report and/or sheets containing rectifications made before this Authority. (see Rule 70.16 and Section 607 of the Administrative Instructions under the PCT).

These annexes consist of a total of 19 sheets.

3. This report contains indications relating to the following items:

- I ☒ Basis of the report
- II ☐ Priority
- III ☐ Non-establishment of report with regard to novelty, inventive step or industrial applicability
- IV ☐ Lack of unity of invention
- V ☒ Reasoned statement under Article 35(2) with regard to novelty, inventive step or industrial applicability; citations and explanations supporting such statement
- VI ☐ Certain documents cited
- VII ☒ Certain defects in the international application
- VIII ☒ Certain observations on the international application

Date of submission of the demand  21 JUNE 2000	Date of completion of this report  30 NOVEMBER 2000
Name and mailing address of the IPEA/US Commissioner of Patents and Trademarks Box PCT Washington, D.C. 20231	Authorized officer  CONSTANTINE HANNAHER
Facsimile No. (703) 305-3230	Telephone No. (703) 308-0956

## INTERNATIONAL PRELIMINARY EXAMINATION REPORT

International application No.

PCT/US99/28038

**I. Basis of the report**

## 1. With regard to the elements of the international application:\*

☐ the international application as originally filed☒ the description:

pages \_\_\_\_\_ (See Attached) \_\_\_\_\_, as originally filed  
pages \_\_\_\_\_, filed with the demand  
pages \_\_\_\_\_, filed with the letter of \_\_\_\_\_

☒ the claims:

pages \_\_\_\_\_ (See Attached) \_\_\_\_\_, as originally filed  
pages \_\_\_\_\_, as amended (together with any statement) under Article 19  
pages \_\_\_\_\_, filed with the demand  
pages \_\_\_\_\_, filed with the letter of \_\_\_\_\_

☒ the drawings:

pages \_\_\_\_\_ (See Attached) \_\_\_\_\_, as originally filed  
pages \_\_\_\_\_, filed with the demand  
pages \_\_\_\_\_, filed with the letter of \_\_\_\_\_

☒ the sequence listing part of the description:

pages \_\_\_\_\_ (See Attached) \_\_\_\_\_, as originally filed  
pages \_\_\_\_\_, filed with the demand  
pages \_\_\_\_\_, filed with the letter of \_\_\_\_\_

## 2. With regard to the language, all the elements marked above were available or furnished to this Authority in the language in which the international application was filed, unless otherwise indicated under this item.

These elements were available or furnished to this Authority in the following language \_\_\_\_\_ which is:

- ☐ the language of a translation furnished for the purposes of international search (under Rule 23.1(b)).  
☐ the language of publication of the international application (under Rule 48.3(b)).  
☐ the language of the translation furnished for the purposes of international preliminary examination (under Rules 55.2 and/or 55.3).

## 3. With regard to any nucleotide and/or amino acid sequence disclosed in the international application, the international preliminary examination was carried out on the basis of the sequence listing:

- ☐ contained in the international application in printed form.  
☐ filed together with the international application in computer readable form.  
☐ furnished subsequently to this Authority in written form.  
☐ furnished subsequently to this Authority in computer readable form.  
☐ The statement that the subsequently furnished written sequence listing does not go beyond the disclosure in the international application as filed has been furnished.  
☐ The statement that the information recorded in computer readable form is identical to the written sequence listing has been furnished.

4. ☒ The amendments have resulted in the cancellation of:

- ☒ the description, pages \_\_\_\_\_ NONE  
☒ the claims, Nos. \_\_\_\_\_ NONE  
☒ the drawings, sheets/fig \_\_\_\_\_ NONE

5. ☒ This report has been drawn as if (some of) the amendments had not been made, since they have been considered to go beyond the disclosure as filed, as indicated in the Supplemental Box (Rule 70.2(c)).\*\*

\* Replacement sheets which have been furnished to the receiving Office in response to an invitation under Article 14 are referred to in this report as "originally filed" and are not annexed to this report since they do not contain amendments (Rules 70.16 and 70.17).

\*\*Any replacement sheet containing such amendments must be referred to under item 1 and annexed to this report.

**V. Reasoned statement under Article 35(2) with regard to novelty, inventive step or industrial applicability; citations and explanations supporting such statement****1. statement**

Novelty (N)	Claims <u>1-13</u>	YES
	Claims <u>NONE</u>	NO
Inventive Step (IS)	Claims <u>NONE</u>	YES
	Claims <u>1-13</u>	NO
Industrial Applicability (IA)	Claims <u>1-13</u>	YES
	Claims <u>NONE</u>	NO

**2. citations and explanations (Rule 70.7)**

Claims 1-11 and 13 lack an inventive step under PCT Article 33(3) as being obvious over Dowben (US005468978A) in view of Jones *et al.* (US005216249A) and Welker *et al.* (US002867727A)

With respect to independent claim 1, Dowben discloses a device 10 (Fig. 1) comprising a mechanism having a layer 11 of boron carbide semiconductor (column 3, lines 14-15) wherein the boron carbide layer is an electrically active part of the device. The device of Dowben is suitable as a heterojunction semiconductor device. Jones *et al.* discloses a neutron detection device (Fig. 3c) comprising a sensing mechanism having a layer 24 of semiconductor which is an electrically active part of the detection device (column 2, lines 48-51). The layer 24 in the device of Jones *et al.* is a polycrystalline diamond doped with boron. Accordingly, it would have been obvious to recognize that the boron carbide layer 11 in the device of Dowben would act as a detector of neutrons in view of the well known behavior of boron and the similarity of polycrystalline diamond (a carbon) to a carbide. Welker *et al.* discloses a neutron detecting device (Fig. 2) comprising a semiconductor body 3 and a monitoring device 7. The sensing mechanism is explained by Welker *et al.* at column 4, lines 32-44. Accordingly, it would have been obvious to modify the device suggested by Dowben and Jones *et al.* to specify that the circuitry connected to the neutron detection device be a monitoring device to record changes in the boron carbide layer 11 as detected by the sensing mechanism as explained by Welker *et al.* With respect to dependent claim 2, the sensing mechanism is inherent to the semiconductor layer containing boron as explained by Welker *et al.* With respect to dependent claim 3, the form of the diode suggested by Dowben, Jones *et al.* and Welker *et al.* is a choice which would have been obvious in view of the variety available of effective performance. With respect to dependent claim 4, the device of Dowben further comprises a layer of silicon 12. With respect to dependent claim 5, the device of Dowben is a heterojunction diode (column 1, line 20). With respect to dependent claims 6 and 7, the dimensions of the layers in the device suggested by Dowben, Jones *et al.* and Welker *et al.* are choices which would have been obvious in view of the desired performance. With respect to dependent claim 8, the number of diodes created by the suggestion of Dowben, Jones *et al.* and Welker *et al.* is a choice which would have been obvious in view of the desired performance. Spectrometry is a known desire for neutron detection, so it would have been obvious to provide a neutron energy absorber to allow neutrons of different energies to reach the individual detectors. With respect to dependent claim 9, the mechanical construction of the device suggested by Dowben, Jones *et al.* and Welker *et al.* is a choice which would have been obvious in view of the desired performance. With respect to dependent claim 10, the utility of  $^{10}\text{B}$  in detecting neutrons is shown by Jones *et al.* and Welker *et al.* The proportion included is a choice which would have been obvious in view of the desired performance. With respect to dependent claim 11, the capability of operating boron carbide semiconductor devices at high temperatures is established by Dowben. With respect to independent claim 13, Dowben, Jones *et al.* and Welker *et al.* suggest a neutron detecting device as explained above. It would have been obvious to use such a device for the intended purpose, detecting neutrons. A method of detecting neutrons in accordance with the disclosures would comprise the steps of positioning the neutron detecting device in a location subject to neutrons, introducing at least one such neutron into the boron carbide layer 11 in the device, and monitoring the interaction with a monitoring device as suggested by element 7 of Welker *et al.*

Claim 12 lacks an inventive step under PCT Article 33(3) as being obvious over the prior art as applied in the (Continued on Supplemental Sheet.)



**VII. Certain defects in the international application**

The following defects in the form or contents of the international application have been noted:

The description is objected to as containing the following defect(s) under PCT Rule 66.2(a)(iii) in the form or contents thereof: under PCT Rule 10.1(a), units of weights and measures shall be expressed in terms of the metric system; the unit "curie" with symbol Ci may qualify for use under PCT Rule 10.1(d) or (e), but the SI derived unit for the derived quantity of radionuclide activity is the becquerel with symbol Bq; the unit of length "micron" as found at page 7, line 8 (now line 10 by the amendment filed with the letter of 13 November 2000) and elsewhere in the international application has not been a unit of the SI since 1967 so its use does not comply with PCT Rule 10.1(a).

Claim 7 is objected to under PCT Rule 66.2(a)(iii) as containing the following defect(s) in the form or contents thereof: the unit of length "micron" as found in claim 7 at line 2 has not been a unit of the SI since 1967 so its use does not comply with PCT Rule 10.1(a).

**VIII. Certain observations on the international application**

The following observations on the clarity of the claims, description, and drawings or on the question whether the claims are fully supported by the description, are made:

The description is objected to under PCT Rule 66.2(a)(v) as lacking clarity under PCT Article 5 because it fails to contain an adequate written description of additional diodes interleaved with a neutron energy absorber. The description is inadequate because: no such *additional* diodes irrespective of any neutron energy absorber is found disclosed.

Claim 8 is objected to as lacking clarity under PCT Rule 66.2(a)(v) because practice of the claimed invention is not adequately described in writing, as required under PCT Rule 5.1(a)(iii), for the reasons set forth in the immediately preceding paragraph. Note that claim 1 does not establish any diode.

The description is objected to under PCT Rule 66.2(a)(v) as lacking clarity under PCT Article 5 because it fails to contain an adequate written description of operation at 500 °C. The description is inadequate because: the specific temperature lacks a specific disclosure.

Claim 11 is objected to as lacking clarity under PCT Rule 66.2(a)(v) because practice of the claimed invention is not adequately described in writing, as required under PCT Rule 5.1(a)(iii), for the reasons set forth in the immediately preceding paragraph.

The description is objected to under PCT Rule 66.2(a)(v) as lacking clarity under PCT Article 5 because it fails to contain an adequate written description of "a sensing mechanism coupled to [the] boron carbide layer". The description is inadequate because: no such coupled sensing mechanism is found disclosed.

Claim 10 is objected to under PCT Rule 66.2(a)(v) as lacking clarity under PCT Article 6 because the claim is indefinite for the following reason(s): the claim recitation does not make it clear that the value of "at least 80% <sup>10</sup>B" refers to the percentage of *the boron atoms found in the* boron carbide layer, not the boron carbide layer itself. See the description at page 8, lines 9-21.

Claim 12 is objected to under PCT Rule 66.2(a)(v) as lacking clarity under PCT Article 6 because the claim is indefinite for the following reason(s): there is no antecedent basis for the recitation of "said detector" at line 3. Line 2 establishes a "neutron detecting device" instead.

Claim 13 is objected to under PCT Rule 66.2(a)(v) as lacking clarity under PCT Article 6 because the claim is indefinite for the following reason(s): there is no antecedent basis for the recitation of "said detector" at line 3. Line 2 establishes a "neutron detecting device" instead.

(Note that the amendment to pages 11 and 12 containing claims 12 and 13 filed with the letter of 13 November 2000 *does not* accomplish the described amendments.)

**Supplemental Box**

(To be used when the space in any of the preceding boxes is not sufficient)

Continuation of: Boxes I - VIII

Sheet 10

**I. BASIS OF REPORT:**

This report has been drawn on the basis of the description,  
page(s) NONE, as originally filed.  
page(s) NONE, filed with the demand.  
and additional amendments:  
Pages 1-10, filed with the letter of 13 November 2000.

This report has been drawn on the basis of the claims,  
page(s) NONE, as originally filed.  
page(s) NONE, as amended under Article 19.  
page(s) NONE, filed with the demand.  
and additional amendments:  
Pages 11-12, filed with the letter of 13 November 2000.

This report has been drawn on the basis of the drawings,  
page(s) NONE, as originally filed.  
page(s) NONE, filed with the demand.  
and additional amendments:  
Pages 1-6, filed with the letter of 13 November 2000.

This report has been drawn on the basis of the sequence listing part of the description:  
page(s) NONE, as originally filed.  
pages(s) NONE, filed with the demand.  
and additional amendments:  
NONE

**5. (Some) amendments are considered to go beyond the disclosure as filed:**

The amendment of the description filed 13 November 2000 is objected to under PCT Article 34(2)(b) because it adds matter into the application that goes beyond the disclosure as originally filed. The added matter which is new is as follows: page 7, line 10 states a measurement for the silicon layer that is 0.001 of that originally stated; page 7, line 13 states a measurement for the boron carbide thickness that is 0.001 of that originally stated; page 7, line 14 states a measurement for the depletion layers that is 0.001 of that originally stated; page 8, line 8 states a measurement for the scale that is 0.001 of that originally stated; claim 7, line 2 states a limitation on the thickness that is 0.001 of that originally stated; it is a fundamental aspect of the SI that a micrometer is three orders of magnitude longer than a nanometer.

**V. 2. REASONED STATEMENTS - CITATIONS AND EXPLANATIONS (Continued):**

immediately preceding paragraph and further in view of NEC Corporation (JP 1-204466 A).

With respect to independent claim 12, Dowben, Jones *et al.* and Welker *et al.* suggest a neutron detecting device as explained above. It would have been obvious to use such a device for the intended purpose, detecting neutrons. A method of detecting neutrons in accordance with the disclosures would comprise the steps of positioning the neutron detecting device in a location subject to neutrons, introducing at least one such neutron into the boron carbide layer 11 in the device, and monitoring the interaction with a monitoring device as suggested by element 7 of Welker *et al.* However, to the extent that the device suggested by Dowben, Jones *et al.*, and Welker *et al.* comprises a sensing mechanism that is inherent to the boron carbide layer 11 as amplified by the explanation of Welker *et al.* as explained above, nevertheless it is known to couple a sensing mechanism to a boron-containing layer in a neutron detecting device as disclosed by the patent document to NEC Corp. (Fig. 1). In view of the equally effective performance in sensing the interaction of an introduced neutron with the boron-containing layer, it would have been obvious to modify the sensing mechanism in the device used in a method of detecting neutrons as suggested by Dowben, Jones *et al.* and Welker *et al.* to be separate as suggested by NEC Corp.

----- NEW CITATIONS -----

NONE

- 1 -

**BORON-CARBIDE SOLID STATE NEUTRON DETECTOR AND**  
**METHOD OF USING SAME**

STATEMENT REGARDING FEDERALLY SPONSORED RESEARCH OR  
DEVELOPMENT

5                   The Board of Regents of the University of Nebraska acknowledges that  
some funding for the research leading to this application was provided by the United  
States Government.

CROSS REFERENCE TO RELATED APPLICATIONS  
Not Applicable.

10                   BACKGROUND OF THE INVENTION

The present invention relates to detection of neutrons. More specifically,  
the present invention relates to a method and device for the efficient detection of neutrons  
that employs a boron-rich semiconductor as an electrically active part of the detection  
device.

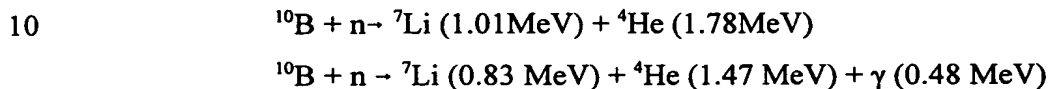
15                   Neutron scattering is an important research method to determine the  
structure of solids and liquids. It is used to understand the forces that act between the  
atoms in these systems and to determine the magnetic behavior of materials as well. The  
research and practical applications cover a broad range of areas, from the basic properties  
of materials to studies of engineering and medical applications.

20                   There are essentially only four elements suitable for forming solid state  
semiconductor neutron detectors – boron (B), cadmium (Cd), gadolinium (Gd) and  
lithium (Li). Lithium semiconductor materials exist ( $\text{LiInS}_2$ ,  $\text{LiInSe}_2$  and  $\text{LiZnP}$ ) but are  
difficult to reliably fabricate into devices and are very difficult materials with which to  
work. Gadolinium conversion layer based silicon (Si) diodes have been fabricated and  
25                   proposed for neutron detection, but are not particularly stable. Cadmium zinc telluride  
has been shown to yield thermal neutron detection and the cadmium neutron capture cross  
section is high, but the neutron capture produces such high energy gamma rays (over 0.5

- 2 -

MeV) that the detectors would have to be large in order to detect these gammas efficiently.

Use of boron with neutron detectors is known both in the scintillator, the gas and the conversion layer varieties. Boron phosphide (BP) heterojunction diodes with silicon were successfully tested as alpha radiation detectors, but failed to work as neutron detectors. Boron carbide ( $B_4C$ ) was successfully used as a neutron detector based upon resistivity changes resulting from increased lithium doping, as were (111) BP wafers. The lithium production in the boron carbide was a result of the following nuclear reactions:



Boron has also been considered as a coating to a silicon diode and a GaAs diode but the maximum efficiency is low (less than 5%).

Existing gas and liquid neutron detectors are much larger and less rugged than solid-state ones could be. However, existing solid state neutron detectors also suffer serious limitations. For example, known boron-doped semiconductors are only a few percent efficient because they contain relatively little boron. Gadolinium, lithium and hydrocarbon conversion layers are all adversely affected by corrosion and high temperatures.

20 Furthermore, known conversion layer devices have low efficiencies, unless multiply stacked, because the range of the reaction products in the material of the conversion layer is generally considerably less than the thickness required for stopping thermal neutrons. Gadolinium conversion layers are an exception - but the neutron - gadolinium reaction results in conversion electrons of relatively low energy (70 keV) compared with the reaction products in the case of neutron capture by boron 10. Cadmium zinc telluride has been shown to yield thermal neutron detection, but the neutron capture produces such high energy gamma rays (over 0.5 MeV) that the detectors must be large to detect these gammas efficiently. Scintillator combinations with photomultipliers or intensified cameras are bulky and heavy and, except for neutron-

- 3 -

detecting scintillating fibers coupled optically to a remote photomultiplier or camera, are intolerant of high temperatures.

Boron and boron compounds, including boron carbide, are also used in neutron absorbing shielding purposes in nuclear reactors and other types of neutron radiation environments. For example, boron carbide can be used with shielding, thermal electric power, or detection of neutrons (by means of the resistivity change not by detection of individual neutrons). However, use of boron carbide to detect neutrons where the boron carbide is an electrically active semiconductor is novel.

#### SUMMARY OF THE INVENTION

10 It is an object of the present invention to provide an inexpensive solid state neutron detector that includes a robust, structurally forgiving boron rich semiconductor.

It is another object of the present invention to provide a boron carbide semiconductor that utilizes its electrical properties as a semiconductor rather than its electrical property of resistance as a means of detecting neutrons or its thermoelectric properties in detecting neutrons.

15 A still further object of the present invention is to provide a detection device that yields high gain.

A further object of the present invention is to provide a detection device that provides real time response.

20 A further object of the present invention is to provide a detection device that is capable of detecting single neutrons.

Yet another object of the present invention is to provide a detection device that has low sensitivity to gamma and other radiation.

25 Still another object of the present invention is to provide a method of detecting neutrons with a detector device having a boron carbide semiconductor.

According to the present invention, the foregoing and other objects are obtained by a detection device having a layer of boron carbide. In the device, the boron carbide layer is an electrically active part of the detection device. The sensing mechanism of the detection device is inherent in the electrically connected, semiconducting boron carbide layer, which provides neutron capture resulting in prompt,

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innately highly amplified, electrical output signals following interception of neutron(s).

Additional objects, advantages, and novel features of the invention will be set forth in part in the description which follows, and in part will become apparent to those skilled in the practice of the invention. The objects and advantages of the invention  
5 may be realized and attained by means of the forms of instrument and the combinations particularly pointed out in the appended claims.

#### BRIEF DESCRIPTION OF THE SEVERAL VIEWS OF THE DRAWINGS

In the accompanying drawings which form a part of the specification and which are to be read in conjunction therewith and in which like reference numerals are  
10 used to indicate like parts in the various views:

Fig. 1 is a schematic representation of a heterojunction diode embodying the present invention;

Fig. 2 is a schematic representation of the test device using the principles of the present invention.

15 Fig. 3 depicts voltage-current characteristics of heterojunction diodes of the present invention;

Fig. 4 depicts count rates of neutrons with insertion of heterojunction diodes of the present invention into a neutron reactor; and

20 Figs. 5 and 6 depict the relationship of ideally attainable neutron detection efficiency as a function of the thickness of the boron-carbide layer of heterojunction diodes of the present invention in the cases of natural 10 Boron abundance and 100% 10 Boron enrichment of the boron carbide layer.

#### DETAILED DESCRIPTION OF THE INVENTION

Referring initially to Fig. 1, a heterojunction diode 10 is shown. This  
25 invention also applies to homojunction diodes and other known semiconductor detection devices, examples of which are provided below. Diode 10 is shown as having a boron carbide boron-carbon alloy semiconductor 12 on a silicon substrate 14. Semiconductor 12 is grown by plasma-enhanced chemical vapor deposition (PECVD). The preferred deposition technique is disclosed in U.S. Patent Nos. 4,957,773 (Spenser, et al.);  
30 5,468,978 (Dowben); 5,658,834 (Dowben), which patents are expressly incorporated by reference herein. A pair of sputter-deposited gold electrodes 16 communicate with





Charge sensitive preamplifier 22, in turn, is connected to a bias voltage input 24 and a single channel analyzer / multichannel scaler 26 which is connected to a computer 28.

In this heterojunction diode, the above reactions lead to dense local ionization of atoms and hence production of electron-hole pairs (at least of order  $5 \times 10^5$  pairs per neutron reaction), many of which are collected due to the applied bias voltage and form a charge pulse which is registered and counted by external circuitry. Such a device was first tested successfully on July 24, 1998 at the nuclear reactor in the VA Hospital in Omaha, Nebraska. This device could be improved in several ways, including  $^{10}\text{B}$ -enrichment (to nearly 100%  $^{10}\text{B}$  from the naturally occurring approximately 19%  $^{10}\text{B}$  found in unenriched boron), increasing the thickness and quality of the boron carbide layer, changes in the electrical configuration and electrical circuitry, and changes in the functional and geometrical configurations.

The deposition of films for the heterojunction diodes (boron-carbon alloy,  $\text{B}_5\text{C}$ , on (111) Si) performed in this test was undertaken in a custom designed parallel plate 13.56 (MHz radio-frequency PECVD reactor used in previous studies). The silicon substrates were doped to  $7 \times 10^{14}/\text{cm}^3$ . The (111) Si substrates surfaces were prepared by  $\text{Ar}^+$  ion sputtering in the plasma reactor. The source molecule gas closo-1,2-dicarbadoecaborane (ortho-carborane,  $\text{C}_2\text{B}_{10}\text{H}_{12}$ ) was used as the source compound for growing the boron carbon alloy.

Typical  $\text{B}_5\text{C}/\text{n}$ -type silicon heterojunctions have been routinely formed by this technique. An example of one such diode device is presented in Fig. 2 with the boron carbide alloy layer of about 1000 nm thick as used as a neutron detector. These devices typically have onsets of 1 eV with very little leakage current (less than  $5 \mu\text{A}$  at  $25^\circ\text{C}$ ) and the boron carbide layer has the p-type character of the undoped PECVD semiconducting boron carbide in this device topology.

The detector area of these heterojunction diodes was about  $1 \text{ cm}^2$ , and wired in a "mesa" geometry. The neutron source was a small TRIGA-type reactor (V.A. Medical Center, Omaha, NE) with a flux of  $1.6 \times 10^6 \text{ n/cm}^2 \cdot \text{s}$  based on calculations for the fission chamber. A heterojunction diode, reversed biased to about 3 V, was wired for pulse counting as shown in Figure 2 and inserted into the reactor. The resulting count

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rates with insertion are plotted in Figure 3. Background and noise counts are in the range of 250 to 300 Hz, and within the reactor, the count rate rises to  $2 \times 10^5$  Hz.

To assure that very little of this count rate is attributable to gamma radiation, the diode was tested against a 100 mCi  $^{137}\text{Cs}$  source for gamma radiation at a distance of 10 cm. The 661 keV gamma rays provided no detectable increase in count rate above background in spite of an expected  $10^6$  gamma rays incident on the diode per second. This is consistent with the expected extremely low gamma-ray sensitivity of such a solid state boron-carbon/silicon semiconductor alloy device, since boron and carbon have low atomic numbers and the boron-rich detectors were made very thin (1000 nm), and the electrically active silicon layer was under 600 nm thick.

Given that almost all counts are attributable to neutrons and that the boron carbide film is about 1000 nm thick, the detection efficiency is thus about 1%. Given that devices can be made with boron carbide of 50 nm to 100 nm in thickness and with depletion layers extending several nanometers, the single (thermal) neutron detection efficiencies are, conservatively, expected to reach 80% and higher in devices which simultaneously have exceedingly low  $\gamma$ -ray sensitivity ( $< 1\%$  detection efficiency for all energies greater than 100 keV and  $< 0.01\%$  for all energies above 0.5 MeV, assured by the use of boron as the dominant atomic species). Since the neutron -  $^{10}\text{B}$  interaction results almost exclusively in the yield of highly ionizing lithium ions and alpha particles of total kinetic energy about 1.5 MeV and the boron atoms form the major species in the active semiconducting regions of the devices, the boron-carbon alloy layer of the detector yields an enormous internal gain (considerably greater than  $10^5$ ) which is essentially noise-free and comparable with the gain of the intensifiers and photomultipliers commonly used in scintillation-based detectors and imagers. By using exclusively  $^{10}\text{B}$  enriched boranes in the PECVD fabrication process, detection efficiency with thinner films can be considerably improved compared with devices whose  $^{10}\text{B}$  content reflects the natural isotopic abundance, about 19%  $^{10}\text{B}$ .

As seen in Figure 2, the electronics demands are minimal compared with those for gadolinium neutron conversion layer-based detectors (which rely on the much smaller 70 keV energetically available for signal generation by the conversion electrons from gadolinium), while ensuring considerably greater efficiency and stability.

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Additionally, the boron-carbon devices can be thinner than 100  $\mu\text{m}$  thick and still achieve nearly 100% thermal neutron detective efficiency. Stacking diodes, interleaved with neutron energy absorbers, to form efficient neutron "calorimeters" or spectrometers is also possible. In combination with boron carbide based high temperature electronics, the boron-carbon based neutron detection systems are expected to be particularly applicable in harsh environments because of the refractory and mechanical performance of boron carbide. The boron-carbon devices may even be fabricated on metal substrates as well as fabricated with spatial resolution that could be on scales smaller than 0.5 nm. There is the possibility of fabricating spatial array detectors, including position sensors for scattering experiments, as well.

High efficiency is achieved because there is a proportionally large amount of boron present in the semiconductor layer. The boron carbide semiconductor has boron of whatever isotope one therefore chooses present in atomic fractions in the order of 80%. This is exceedingly rich in boron compared with any other suitable semiconductor. Because the density of boron atoms in the material is so high, the boron-rich layer can be quite thin and still contain enough boron atoms per unit area to be able to detect the neutrons very efficiently. In naturally occurring boron there is close to 20% of the boron atoms which are  $^{10}\text{B}$  atoms which are the isotopes which interact strongly with neutrons to give the reactions provided above. It is certainly possible to increase the fraction of boron that is  $^{10}\text{B}$  from natural abundance to about 95% or higher. This enrichment would result in  $^{10}\text{B}$  atoms accounting for a fraction, about 80% or higher, of all atoms in the semiconductor boron carbide layer. Hence, if material enriched in  $^{10}\text{B}$  is used rather than just the naturally occurring isotope ratio of  $^{10}\text{B}$ , the efficiency increases even further.

Another important issue for efficiency is not just the reaction of the neutron with boron, but the ability to detect the reaction. By incorporating the boron atoms in an electrically active semiconductor where the lithium ion and the alpha particle can cause dense ionization of other atoms, many electron-hole pairs can be created by ionization of the atoms, and the electric fields that can be applied across the boron carbide layer can sweep out a large fraction of the electron-hole pairs. Thus, there are three aspects to efficiency. The first is  $^{10}\text{B}$  being present in large number density. The second

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being that the reaction of  $^{10}\text{B}$  with neutrons results in ions which very efficiently ionize atoms in the surrounding in an electrically active semiconductor where the charge can be swept out efficiently. The third aspect of efficiency is that  $^{10}\text{B}$  results in ions which have such a large energy that they can produce very large numbers of detectable electron hole  
 5 pairs. The reactions which occur between neutrons and the other elements which give probable neutron interactions don't result in reaction products which are as readily detectable or detectable to give such large signals. Boron is unique.

Another point concerns detection devices having conversion layers containing boron. Neutron capture by boron generates the alpha particle and the lithium  
 10 ion which can only travel a very limited distance. If conversion layer contained enough boron atoms to cause capture of a sufficient fraction of neutrons, then the layer will be so thick that the lithium and the alpha particles in some cases will not get out of the boron layer and, therefore, will not generate signals that are readily detectable. This is a severe defect compared with the boron carbide semiconductor devices of the present invention.

15 This invention can be used in various forms of solid-state neutron detectors presenting entrance detecting areas of order  $\mu\text{m}^2$  to  $\text{m}^2$ . These detectors are capable of being implemented with very thin detecting and electrically active regions ( $\leq 1 \mu\text{m}$  minimum effective electrical thickness), with very low mass per unit detecting area, with efficiencies ranging up to nearly 100% even for single neutrons, with real-time  
 20 response, with high spatial resolution ( $\leq 1 \mu\text{m}$  minimum), and with high temporal resolution. Of course, implementation may not always need to, or be able to, employ each of these attributes. Voltage and power needs are slight, as are charge pulse processing requirements.

Although the invention is described above as relating to heterojunction  
 25 diodes, it is to be understood that the invention can be implemented in a large number of other ways, including homojunction diodes; p-i-n diodes; metal-semiconductor-metal, Schottky and other diodes; transistors; diode and transistor arrays; charge-induced devices (CID) and CID arrays; charge-coupled devices (CCD) and CCD arrays; solid-state neutron-detecting analogs of "photomultipliers"; neutron semiconductor avalanche  
 30 devices; position-sensitive detectors, including those relying on charge subdivision or sensing and on current subdivision and those having capacitive or resistive means of

- 10 -

doing so; semiconductor drift detectors or semiconductor drift chambers; stacked series of one or more of the above detector types which are configured to serve as neutron energy spectrometers; individual or stacked series of one or more of the above detector types which also, or alternatively, serve as dosimeters. The dosimeters can be capable  
5 of yielding both real-time and cumulative dosimetry information once or many times, completely nondestructively of the dosimetry information contained in the detectors.

The range of applicability of the present invention includes: medical radiation dosimetry; detecting nuclear material; anti-terrorism and anti-smuggling devices; monitoring of nuclear reactors, of nuclear storage units and facilities, and of  
10 of nuclear weapons, weapons storage and weapons shipment; life science materials and physical sciences scattering experiments; monitoring of neutron sources; calibration of neutron flux; personnel and environmental radiation protection; radiation protection at high energy radiation facilities, including medical x-ray facilities (high energy ones); neutron cancer therapy; profiling of medical, therapeutic, research and other neutron  
15 beams; comet, planetary and other space exploration.

From the foregoing, it will be seen that this invention is one well adapted to attain all the ends and objects herein above set forth together with other advantages which are obvious and which are inherent to the structure. It will be understood that certain features and subcombinations are of utility and may be employed without  
20 reference to other features and subcombinations. This is contemplated by and is within the scope of the claims.

Since many possible embodiments may be made of the invention without departing from the scope thereof, it is to be understood that all matter herein set forth or shown in the accompanying drawings is to be interpreted as illustrative and not in a  
25 limiting sense.

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Having thus described the invention, what is claimed is:

1. A neutron detection device, said device comprising: a sensing mechanism, said sensing mechanism having a layer of boron carbide semiconductor wherein the boron carbide layer is an electrically active part of said detection device; and a monitoring device, wherein said monitoring device records said changes in said boron carbide layer detected by said sensing mechanism.
2. The device of claim 1, wherein said sensing mechanism is inherent in said boron carbide semiconductor layer and results in a prompt, innately highly amplified, electrical output following capture of a single neutron.
3. The device of claim 2, wherein said device is a homojunction diode.
4. The device of claim 1, further comprising a layer of silicon communicating with said layer of boron carbide.
5. The device of claim 4, wherein said device is a heterojunction diode.
6. The device of claim 1, wherein the thickness of said boron carbide layer is about 1000 nm.
7. The device of claim 5, wherein the thickness of said silicon layer is less than 600 nm.
8. The device of claim 1, further comprising at least two diodes interleaved with a neutron energy absorber.
9. The device of claim 1, wherein said boron carbide layer is fabricated on a metal substrate.
10. The device of claim 1, wherein said boron carbide layer contains at least 80%  $^{10}\text{B}$ .
11. The device of claim 1, wherein said device is capable of operating at 500 °C.
12. A method of detecting neutrons, said method comprising: positioning a neutron detecting device in a location to allow said device to intercept a stream of neutrons, said detector comprising a layer of boron carbide wherein said boron carbide layer is an electrically active part of said device, and a sensing mechanism coupled to said boron carbide layer; introducing at least one neutron traveling in a direction to be intercepted by the boron carbide layer; and monitoring the interaction of the neutron with

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the boron carbide semiconductor; wherein said sensing mechanism detects changes in said boron carbide layer caused by the interception of neutrons.

13. A method of detecting neutrons, said method comprising: positioning a neutron detecting device in a location to allow said device to intercept a stream of neutrons, said detector comprising a layer of boron carbide wherein said boron carbide layer is an electrically active part of said device, and a sensing mechanism inherent to said boron carbide layer; introducing at least one neutron traveling in a direction to be intercepted by the boron carbide layer; and monitoring the interaction of the neutron with the boron carbide semiconductor; wherein said sensing mechanism detects changes in said boron carbide layer caused by the interception of neutrons.

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**ABSTRACT OF THE DISCLOSURE**

A boron carbide solid state neutron detector and method of using the detector is disclosed, wherein the detector includes a layer of boron carbide wherein the boron carbide layer is an electrically active part of the detection device, a sensing mechanism inherent to said boron carbide layer, wherein the sensing mechanism detects changes in the boron carbide layer caused by the interception of neutrons and a monitoring device coupled to the sensing mechanism.



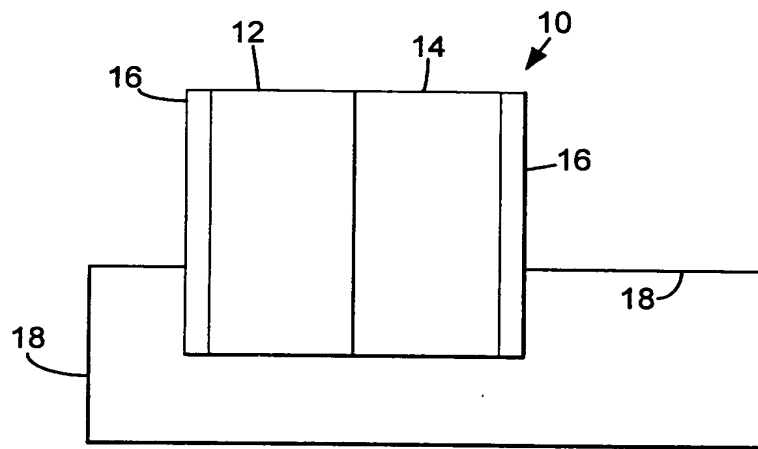
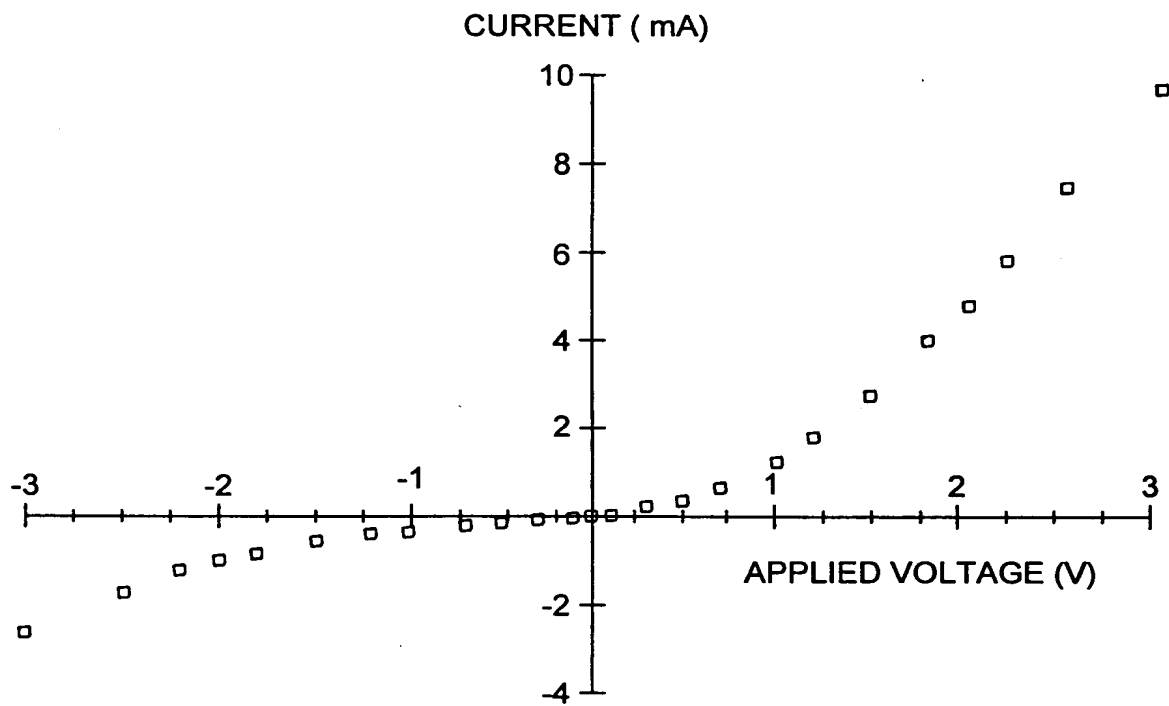


FIG.1.

FIG.3.



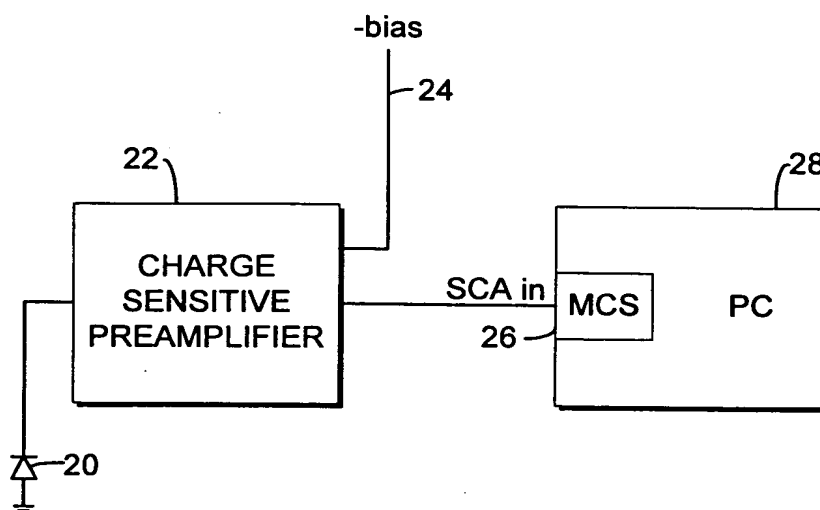
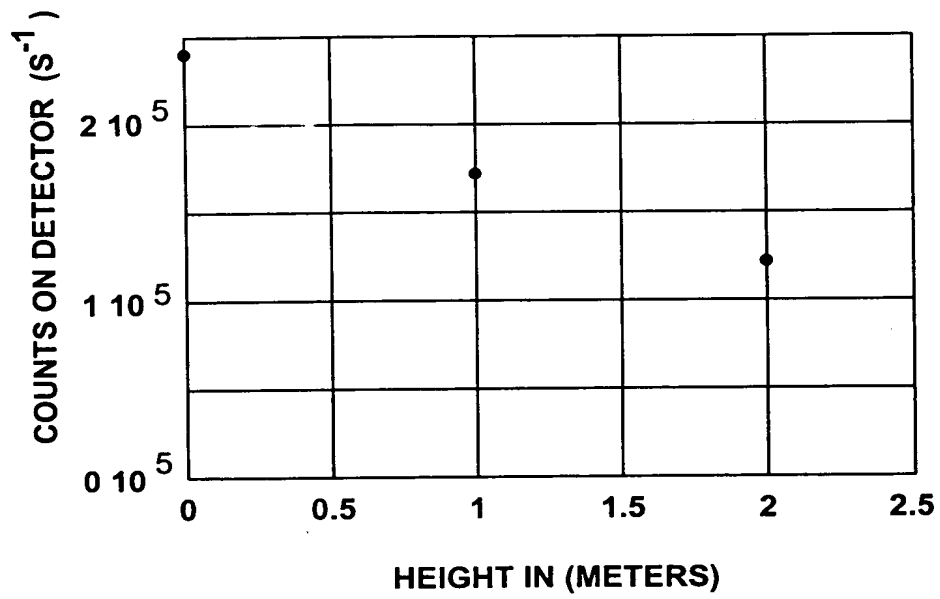
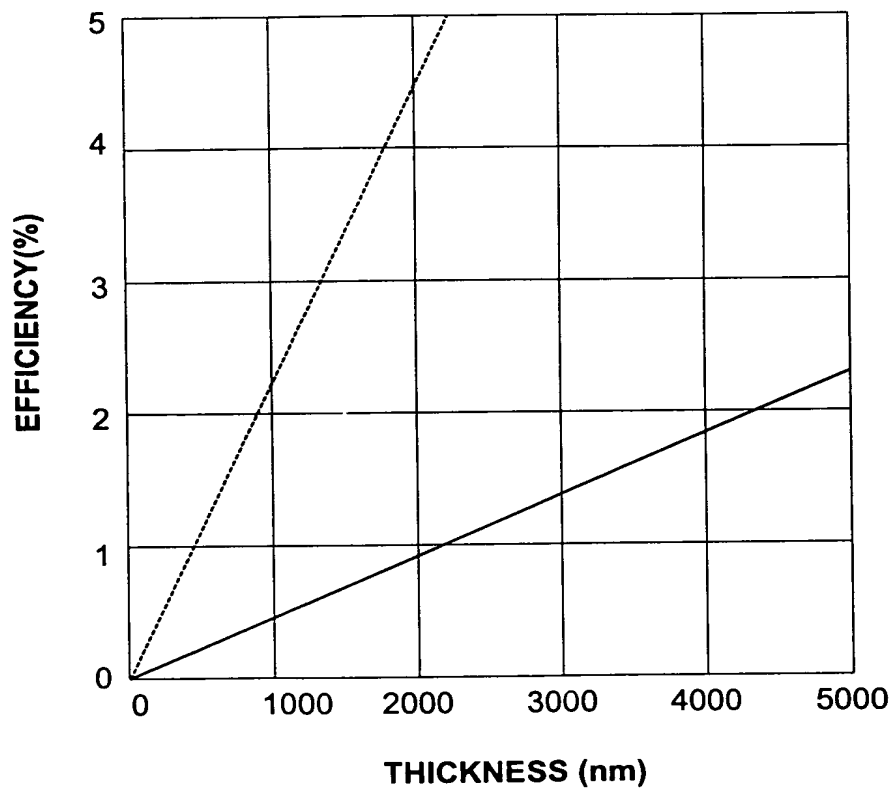
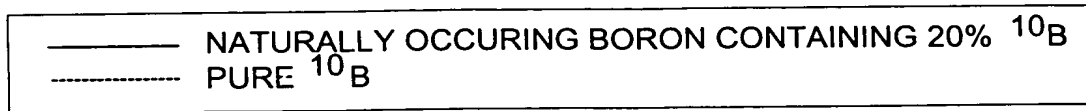


FIG.2.



**FIG. 4.**

FIG. 5.



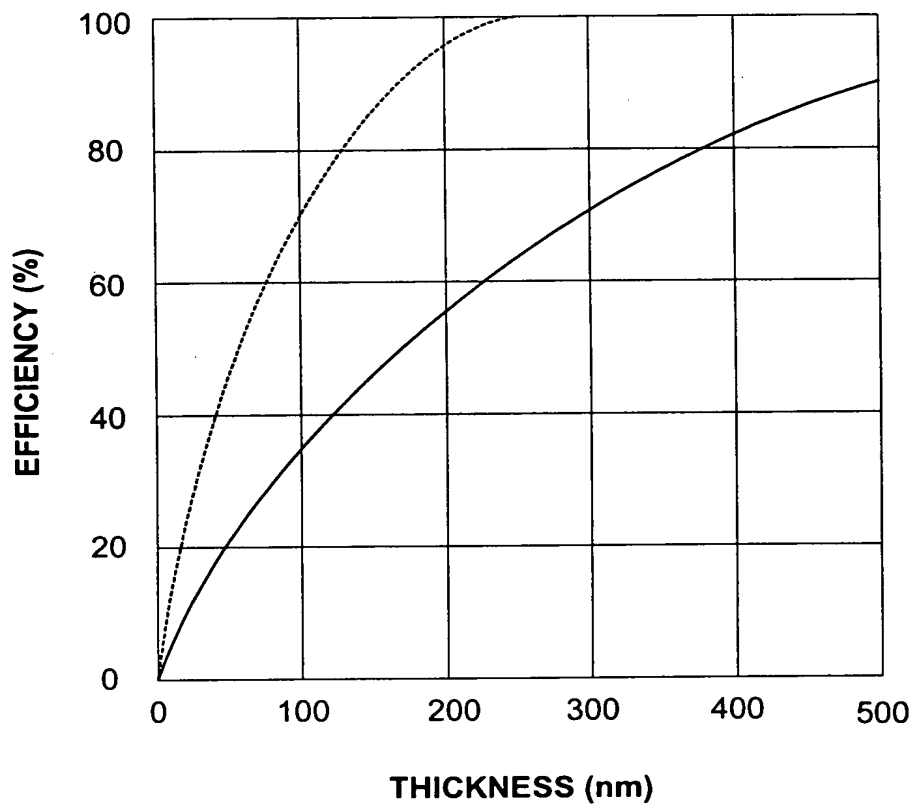
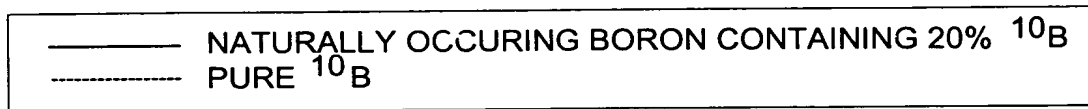


FIG. 6.